



Argonne
NATIONAL
LABORATORY

... for a brighter future



U.S. Department
of Energy



THE UNIVERSITY OF
CHICAGO

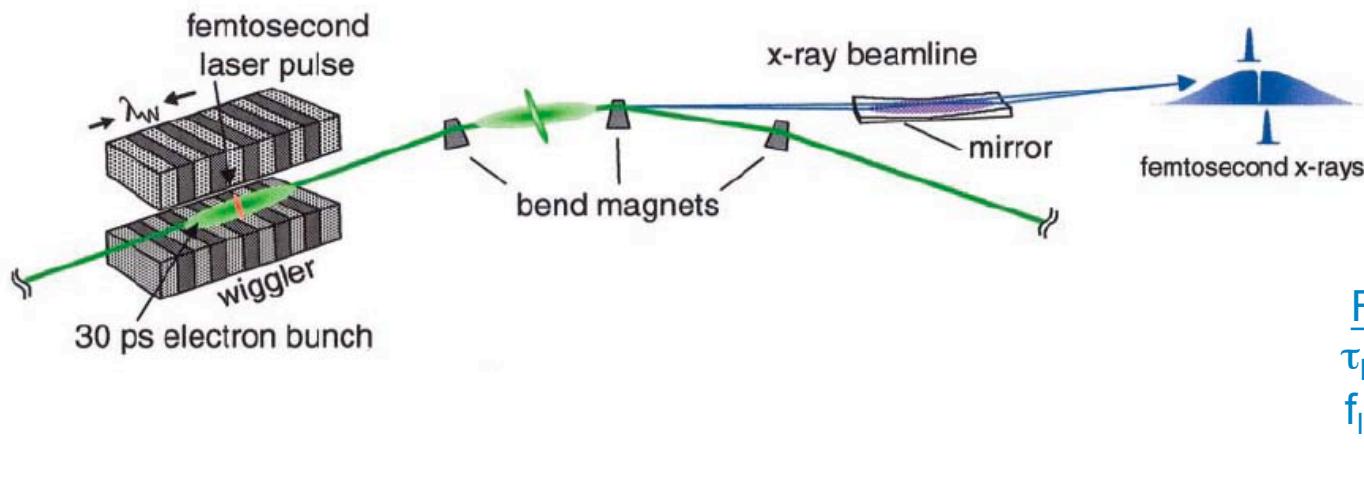


A U.S. Department of Energy laboratory
managed by The University of Chicago

Why do we want 1-ps x rays?

Linda Young
SPX Mini workshop
Argonne National Laboratory
February 15, 2008

Synchrotron-based tunable x-ray sources



Flux reduction

$$\tau_{\text{laser}}/\tau_{\text{elec}} \sim 10^{-3}$$
$$f_{\text{laser}}/f_{\text{elec}} \sim 10^{-3}$$
$$\epsilon_{\text{defl}} \sim 0.1$$

Concept: Zholents and Zolotorev, Phys. Rev. Lett. **76**, 916 (1996).

Demonstration bend magnet at ALS: 100 fs, 5/pulse, $\Delta E/E \sim 0.1\%$, 1 kHz

R. W. Schoenlein et al., Science **287**, 223 (2000).

Demo undulator at BESSY II: 100 fs, 1000/pulse, $\Delta E/E \sim 0.1\%$, 1 kHz, 41-8.8 Å

S. Khan et al., Phys. Rev. Lett. **97**, 074801 (2006).

Demo undulator at SLS: 140 fs, 200/pulse, $\Delta E/E \sim 0.1\%$, 1 kHz, 2.5-1 Å

P. Beaud et al., Phys. Rev. Lett. **99**, 174801 (2007).

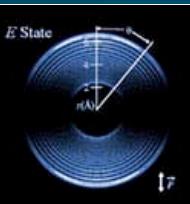
Ultrafast x-ray science workshops up to 2006

**Workshop on New Opportunities in
Ultrafast Science using X-rays**
April 14-17, 2002, in Napa CA



**Corsica Summer School 2003
Ultrafast X-ray Science**

**ULTRAFAST
X-RAYS 2004**



**Ultrafast Science with X-rays and Electrons
Montreux, Switzerland, 9-12 April 2003**



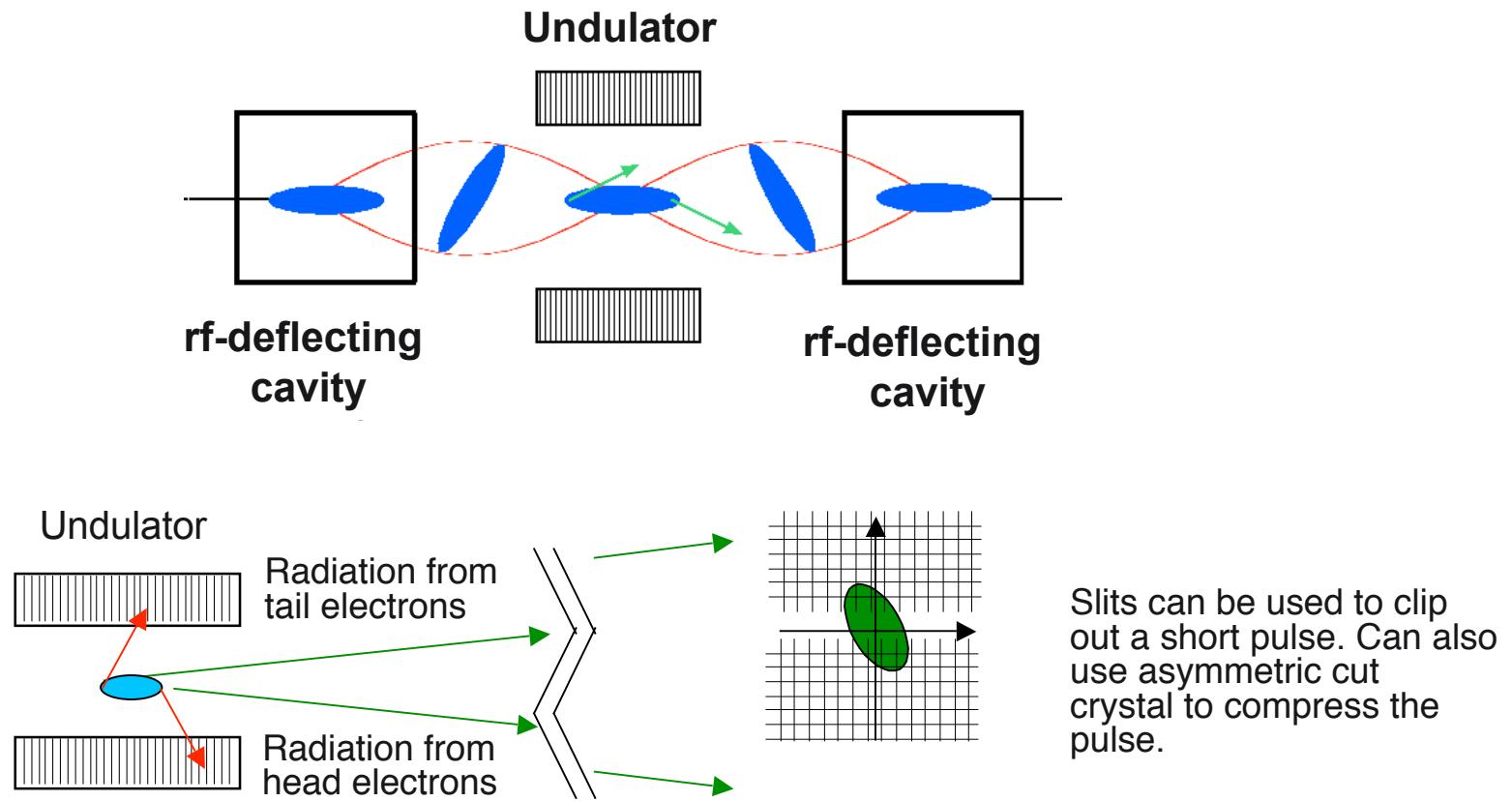
**Time Domain Science Using X-ray Techniques
Aug 29-Sep 1, 2004, Lake Geneva**



**Cornell University June 2006
ERL X-ray Science Workshops**

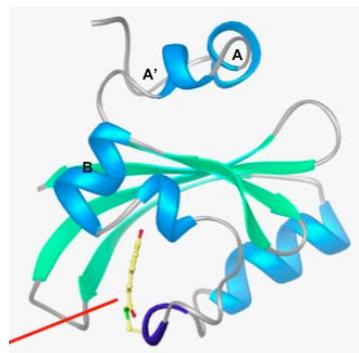


APS short pulse x-ray project: 1-ps, 10^6 /pulse

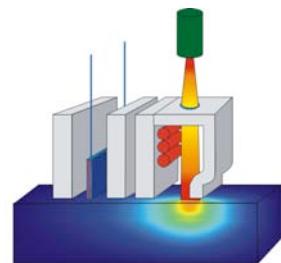
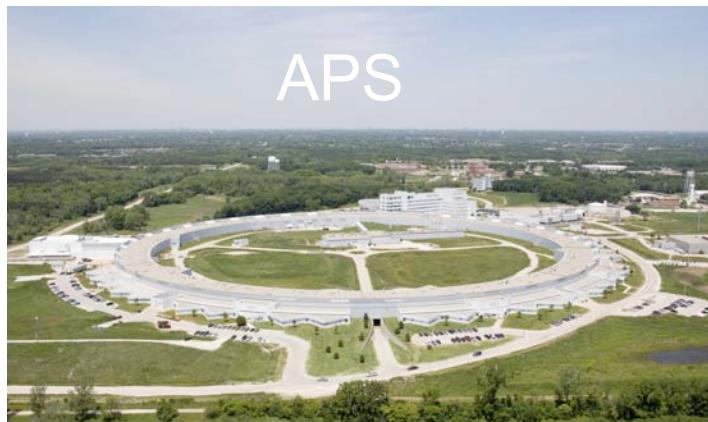


Concept: A. Zholents, P. Heimann, M. Zolotorev, J. Byrd, NIM **A425** (1999).
Simulation for APS: M. Borland, PRSTAB **8**, 074001 (2005).
Cavity design/machine studies: A. Nassiri, V. Sajaev, K. Harkay ...

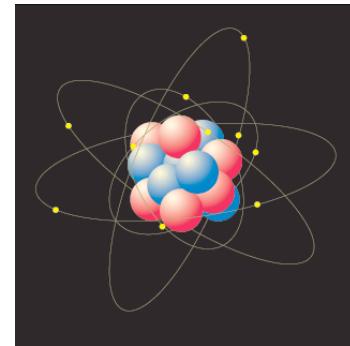
Excitement - transcends disciplines



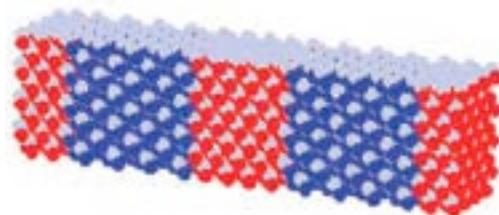
Biology



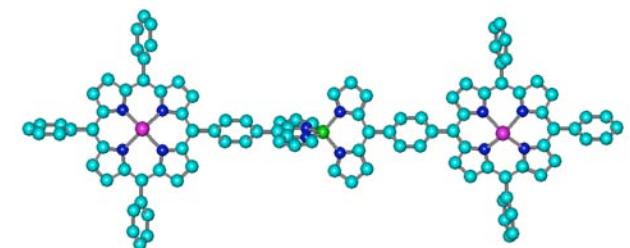
Materials Science



AMO



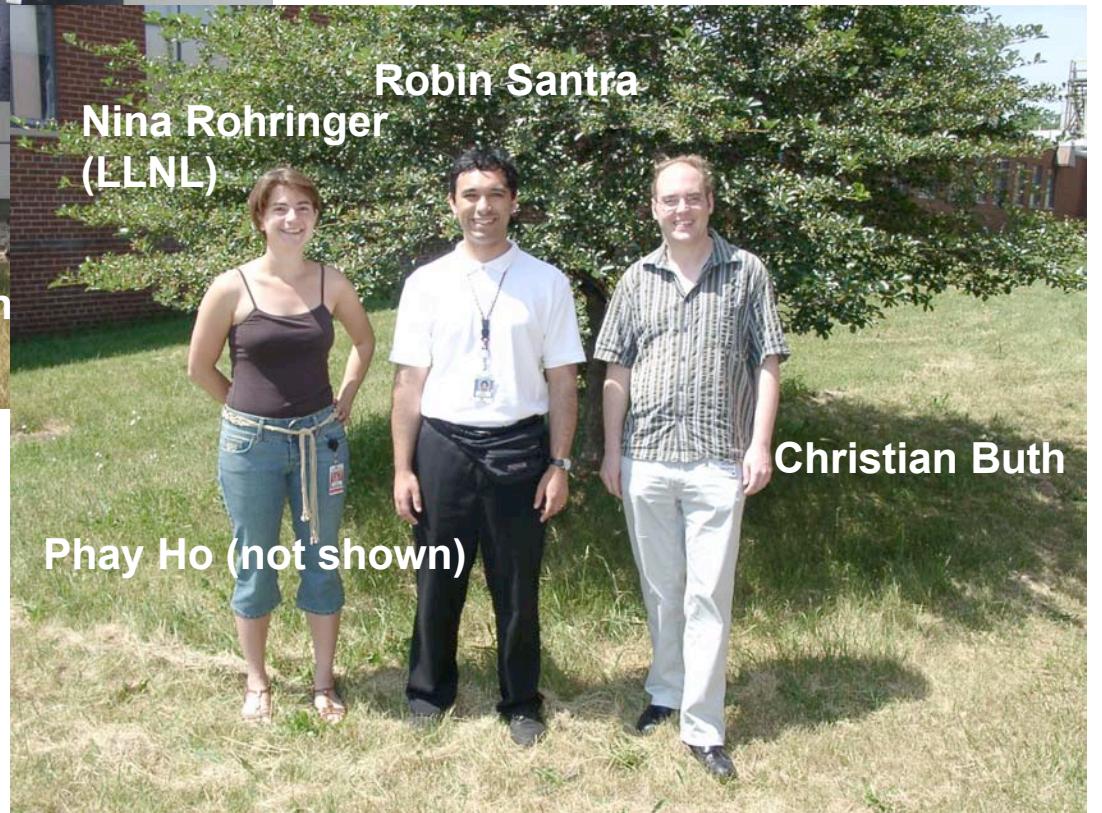
Condensed Matter



Chemistry



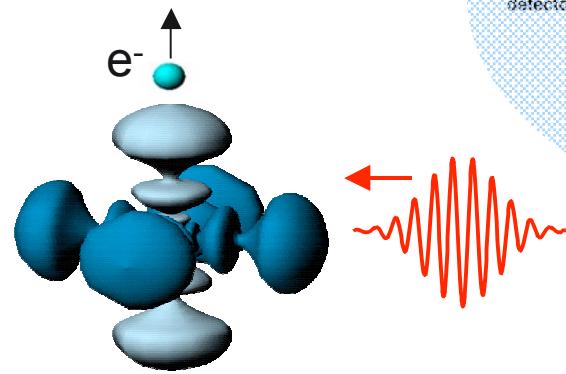
AMO Physics



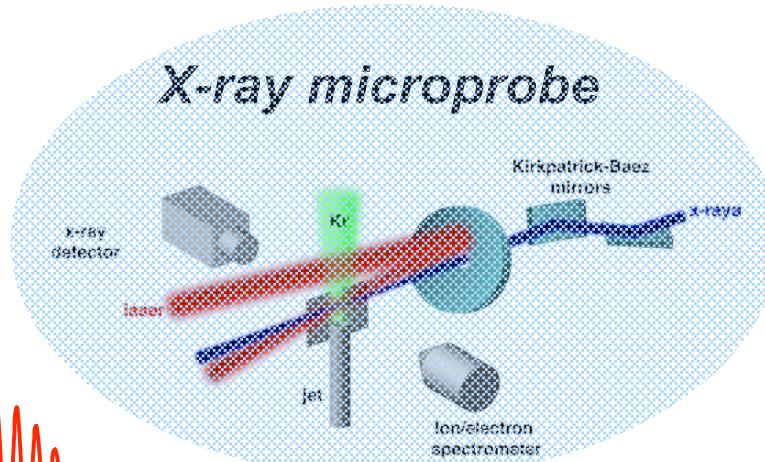
Collaborators:

D. L. Ederer (Tulane)
S. T. Pratt (Argonne)
E. C. Landahl (Argonne/APS)
E. Dufresne (Argonne/APS)
D. A. Arms (Argonne/APS)

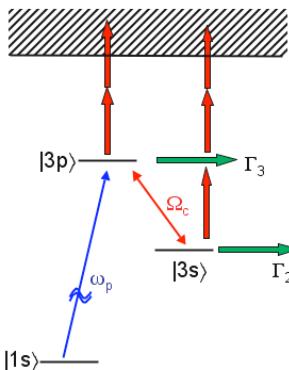
Strong-field control of x-ray processes



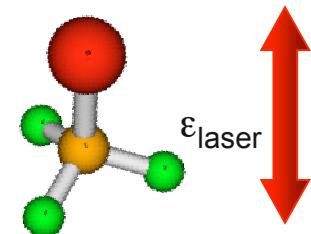
**Orbital alignment in
ultrafast field ionization**
 $10^{14}\text{-}10^{15} \text{ W/cm}^2$



Tunable, polz'd x-rays probe
atoms & molecules at
 $10^{12}\text{-}10^{14} \text{ W/cm}^2$

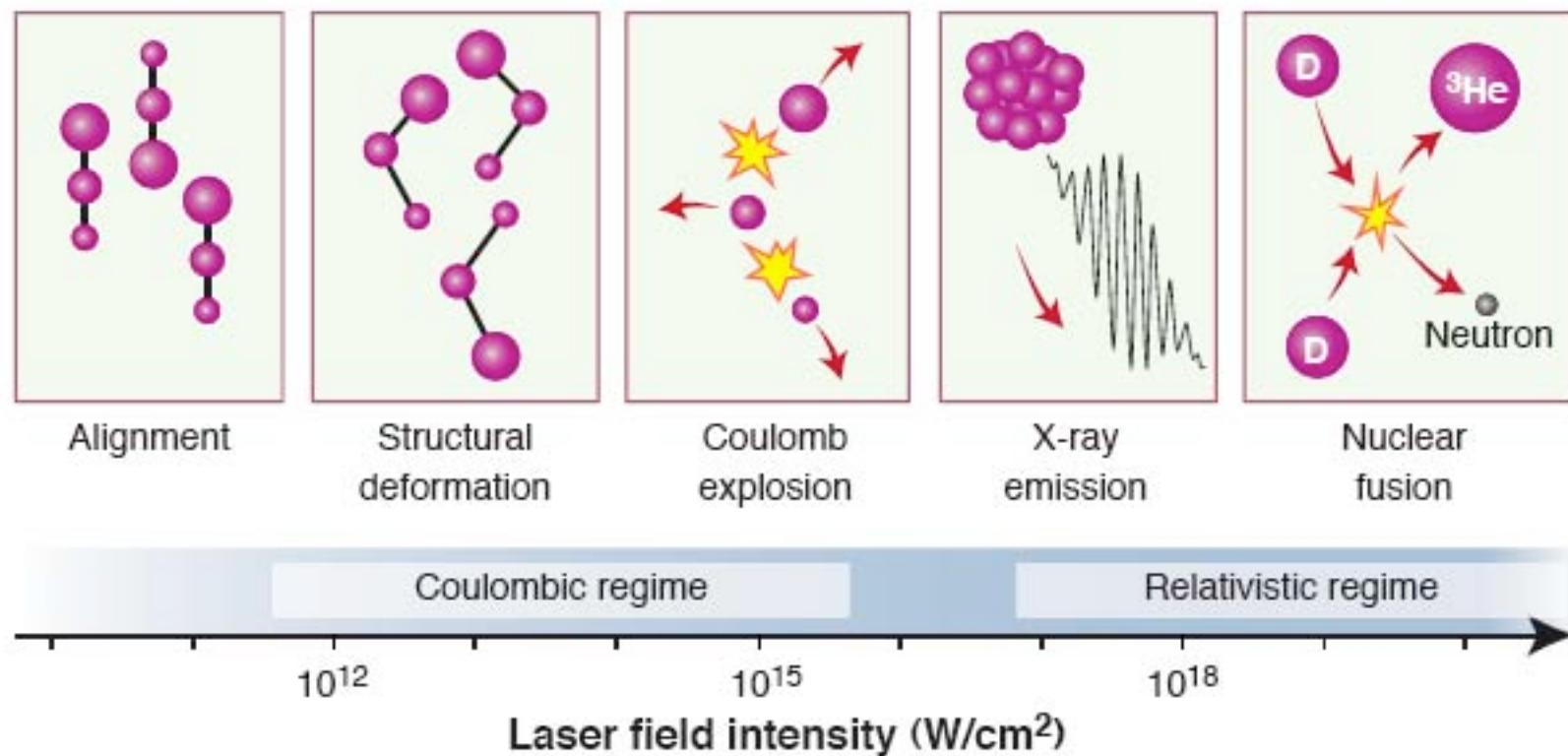


**Electromagnetically induced transparency
for x-rays**
 $\sim 10^{13} \text{ W/cm}^2$



**X-ray absorption by
laser-aligned molecules**
 $\sim 10^{12} \text{ W/cm}^2$

Beyond alignment: behavior of molecules in strong laser fields

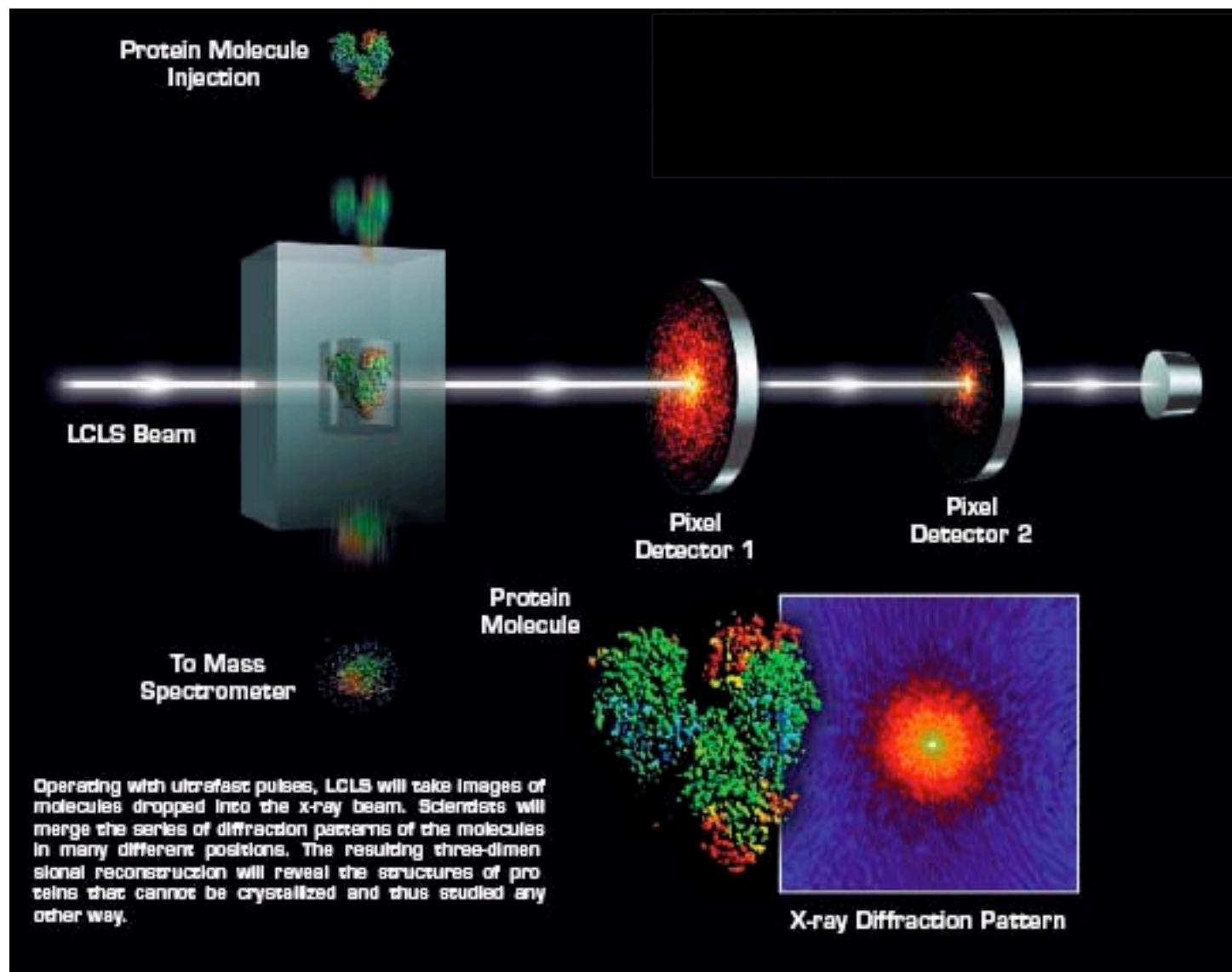


Yamanouchi, Science (2001)

Excitement

*Coherent diffractive imaging of aligned individual molecules
with joint Ångstrom/picosecond resolution
with phase retrieval/inversion → molecular movies*

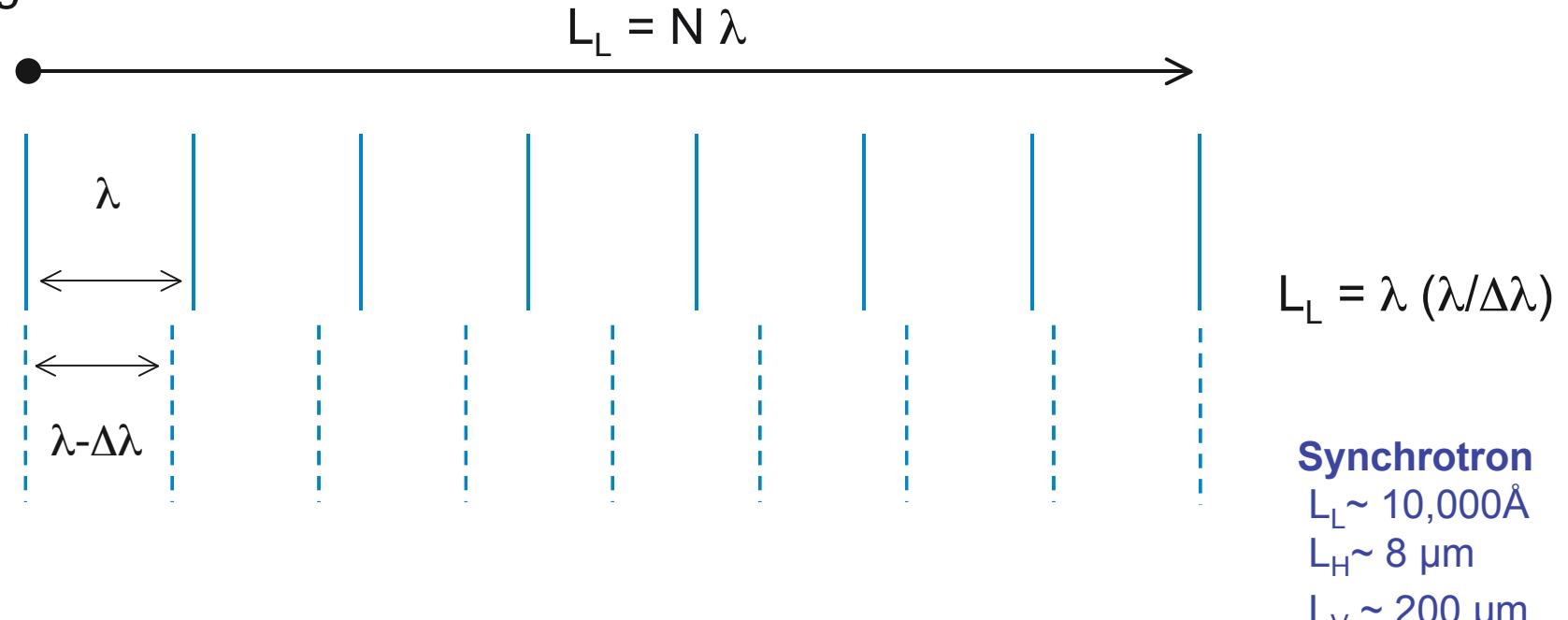
“Serial crystallography” with intense x-rays - LCLS



Simplified with aligned molecules - Spence & Doak PRL(04)

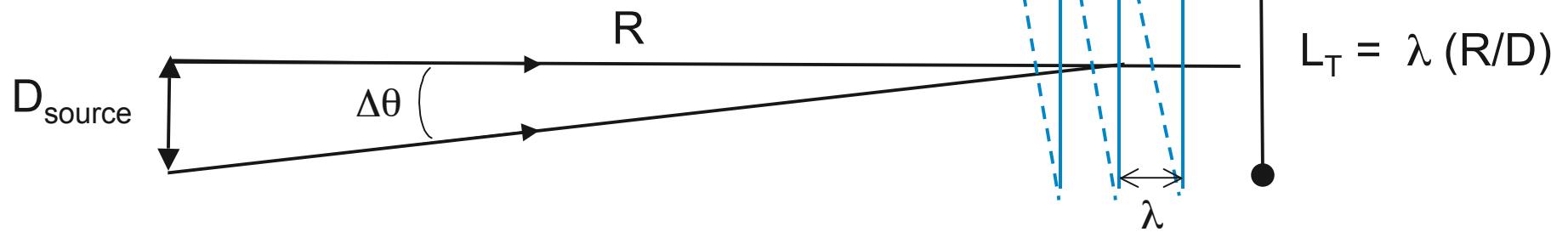
Coherence properties

Longitudinal coherence

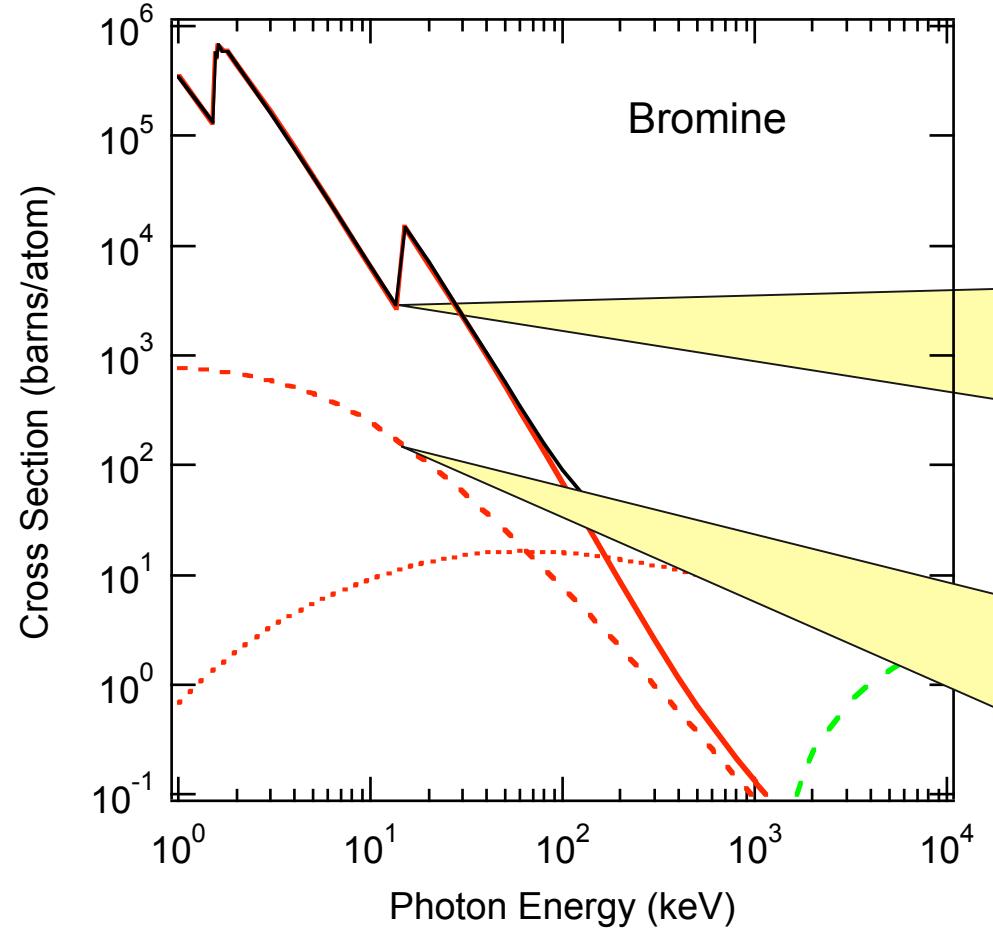


Synchrotron
 $L_L \sim 10,000 \text{ \AA}$
 $L_H \sim 8 \mu\text{m}$
 $L_V \sim 200 \mu\text{m}$

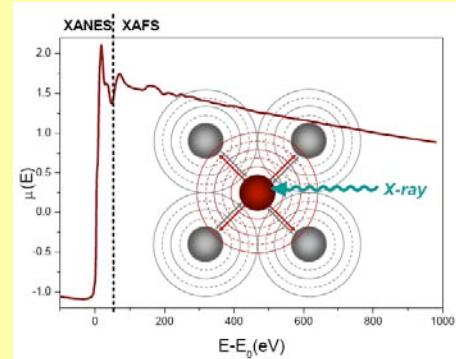
Transverse coherence



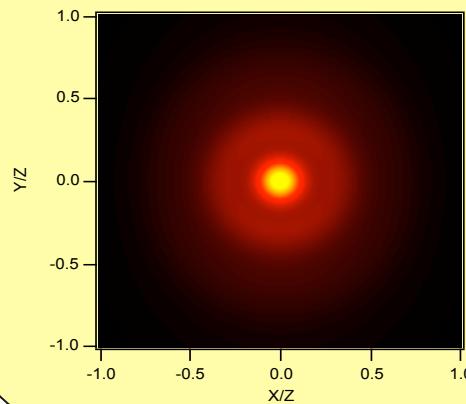
X-ray processes: absorption and scattering



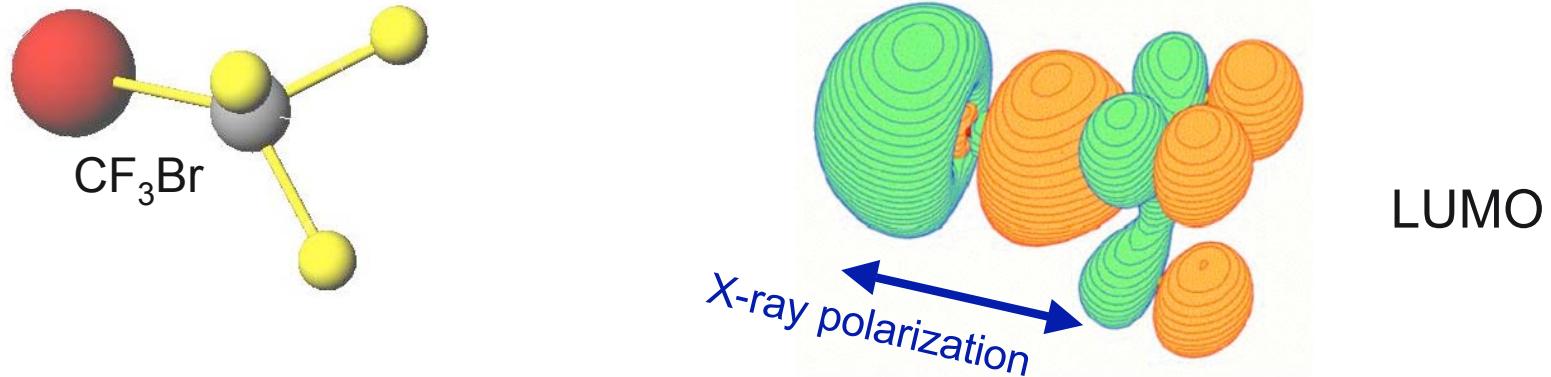
Photoabsorption



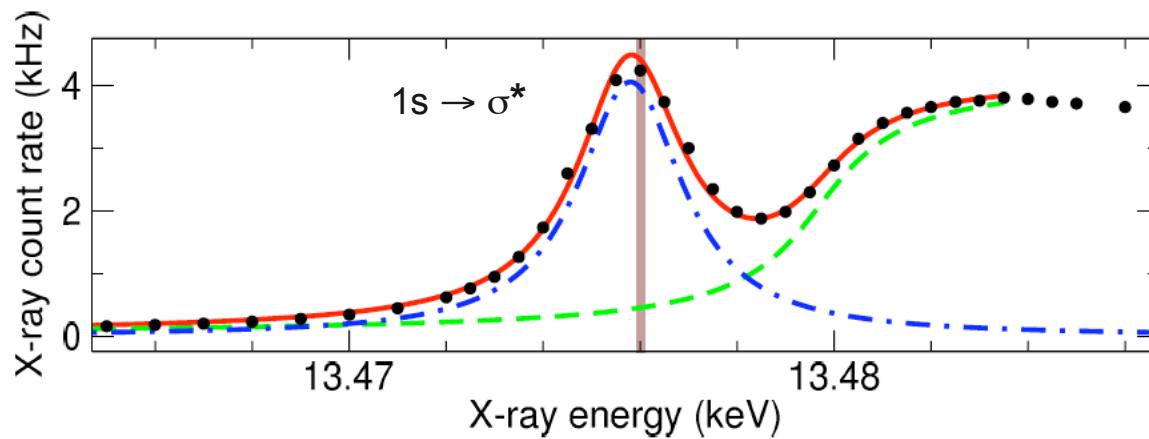
Elastic scattering



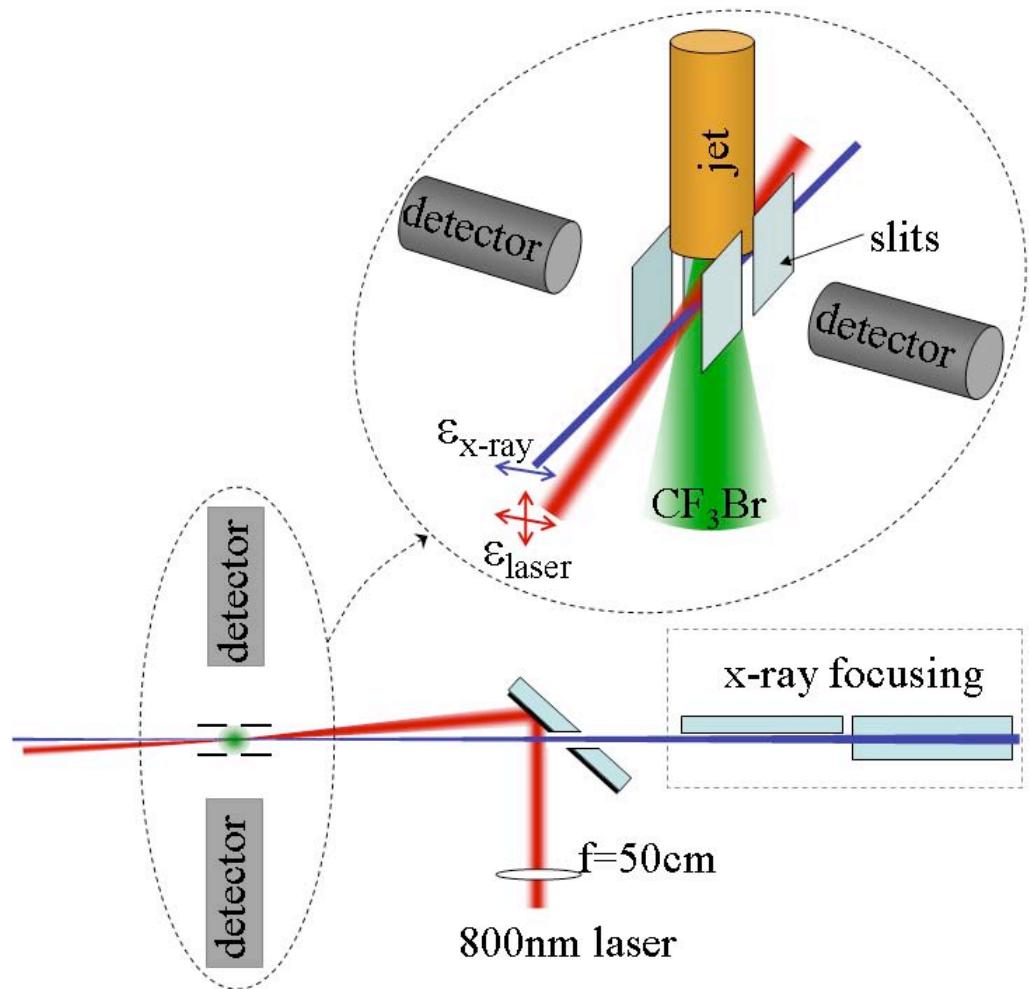
Laser-controlled near-edge absorption: align CF_3Br & polzn



LUMO: σ^* orbital with large Br $4p_z$ character;
resonant x-ray absorption by Br K-shell
electron probes only this Br $4p_z$ component



X-ray absorption by a laser-aligned molecule: CF_3Br



Laser:

1.9 mJ, 95 ps, 800 nm, 40 μm

X-rays:

10^6 photons, 120 ps, 13.4 keV, 10 μm

Detection x-ray fluorescence ($Br K\alpha$):

1.2 mm viewed overlap region

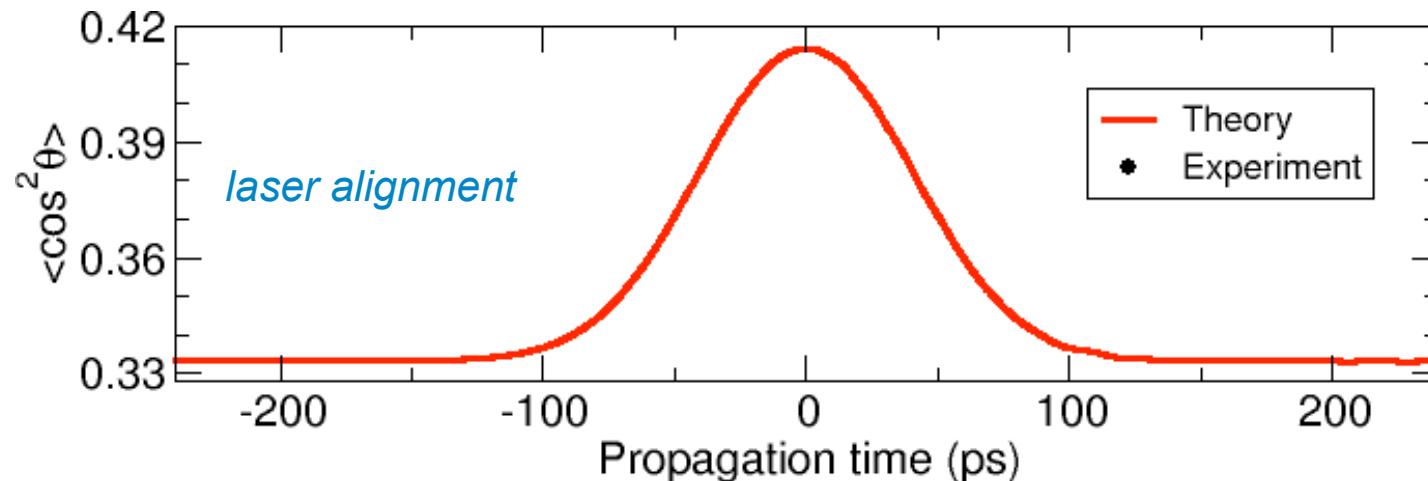
$0.85 \times 10^{12} \text{ W/cm}^2$ peak laser intensity

Sample:

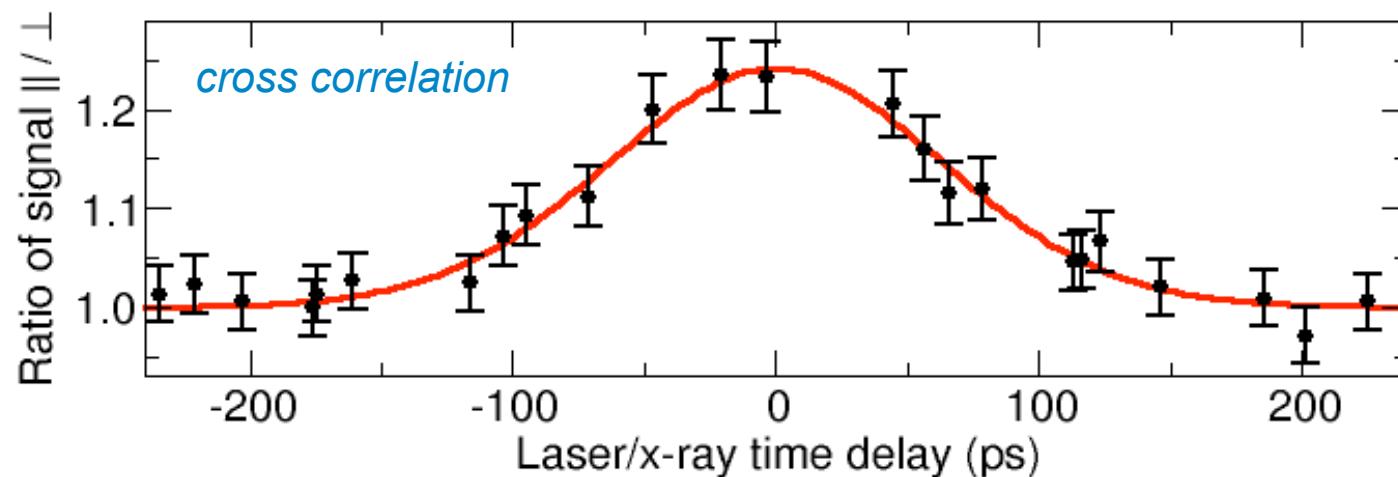
Supersonic expansion: CF_3Br/He

X-ray absorption of a laser aligned molecule: CF_3Br

95 ps laser pulse, $8.5 \times 10^{11} W/cm^2$, 20 K rotational temperature, 122 ± 18 ps x-ray pulse

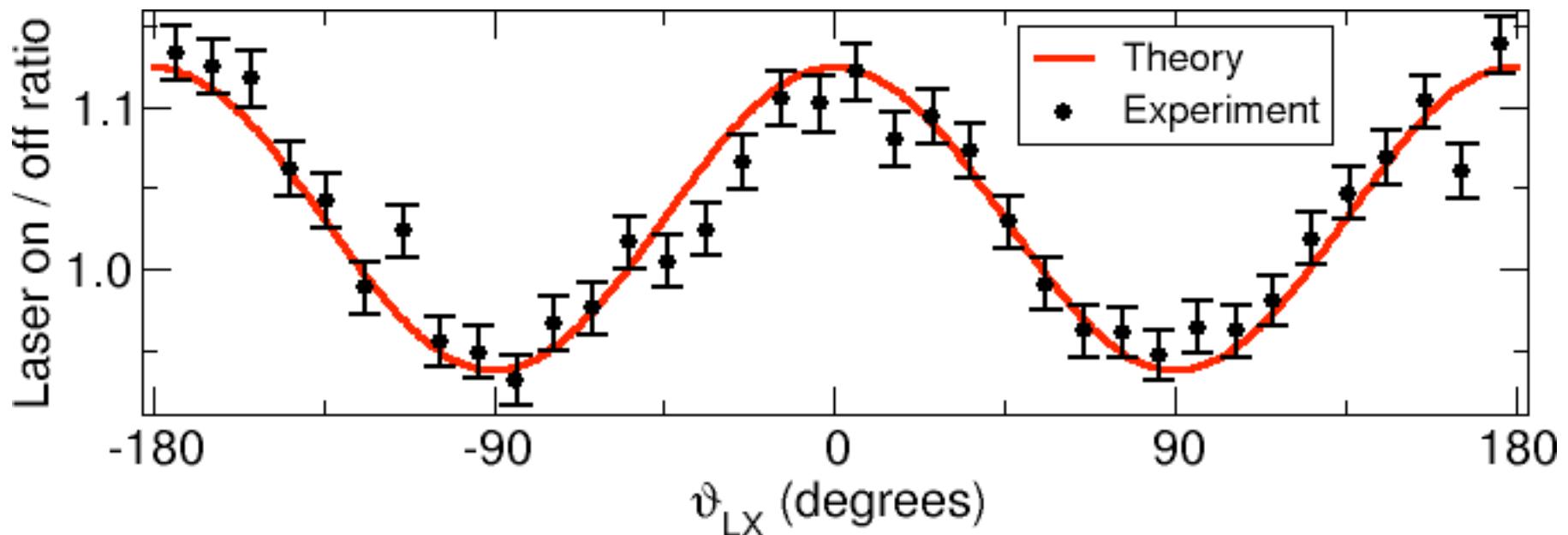


235 ps ground state rotational period, but 28 ps average period at 20 K

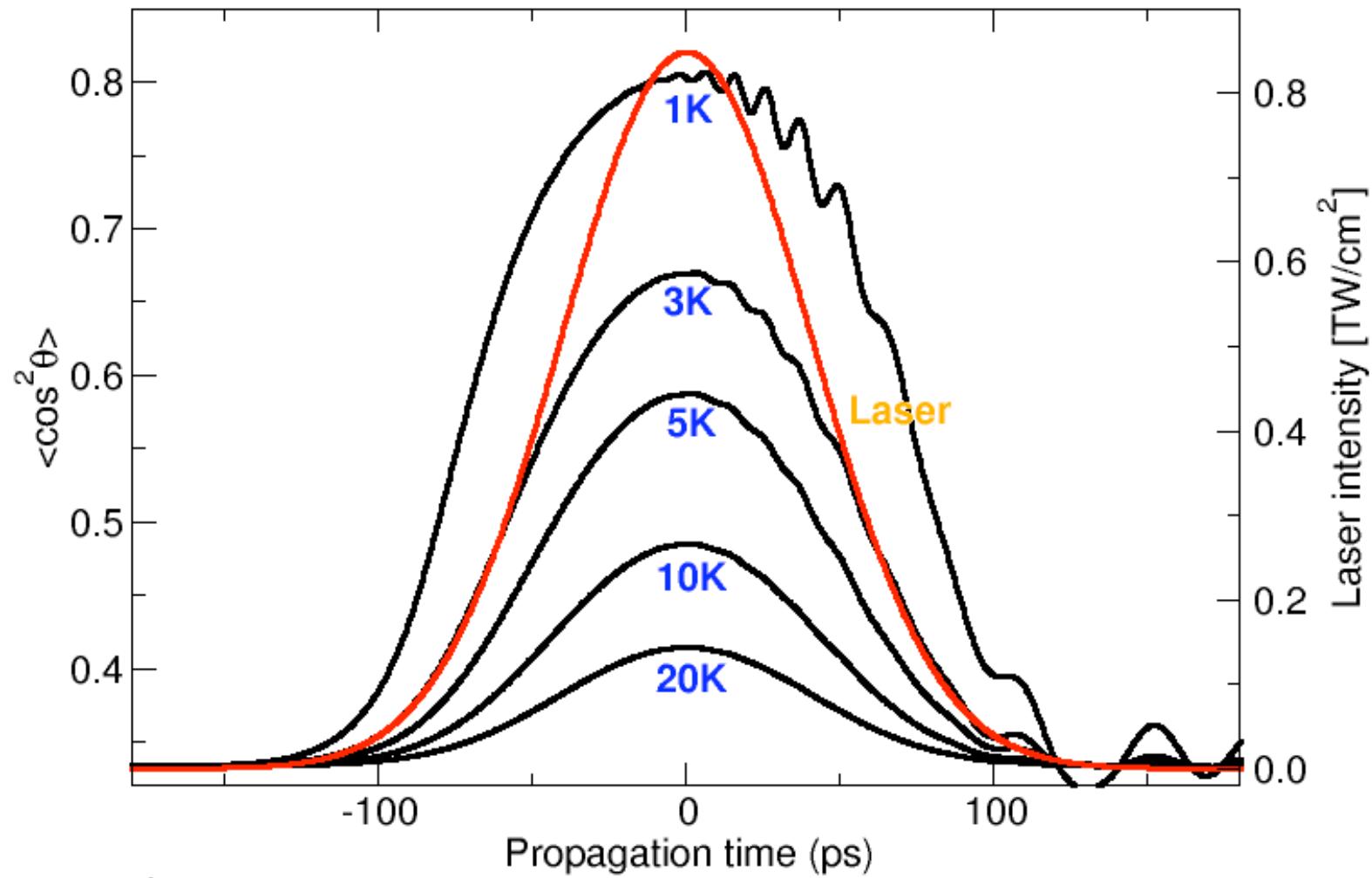


X-ray absorption of a laser aligned molecule: CF_3Br

laser-on/laser-off ratio vs. angle between laser and x-ray polarizations

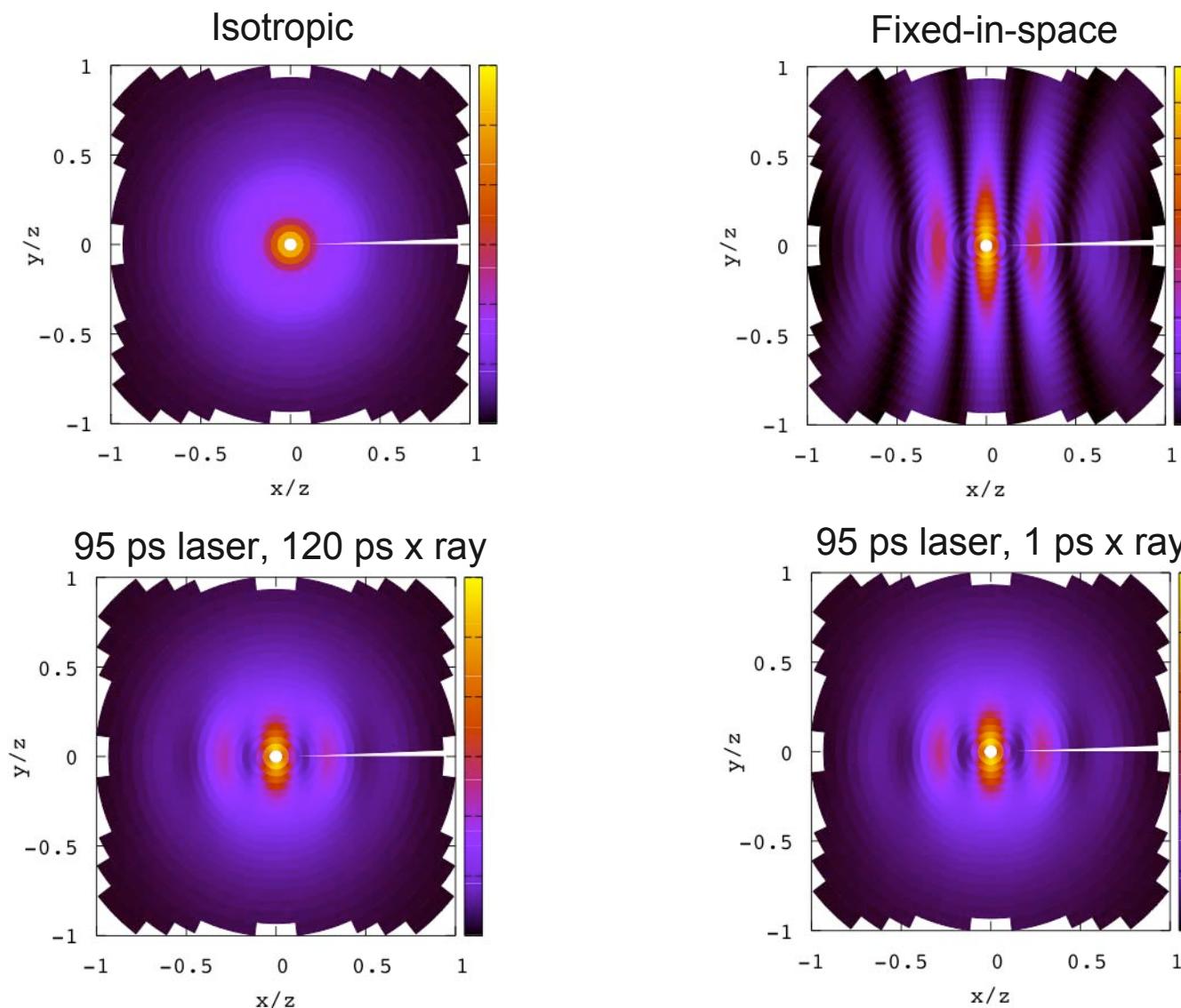


Future 1: Stronger alignment at lower rotational temperatures

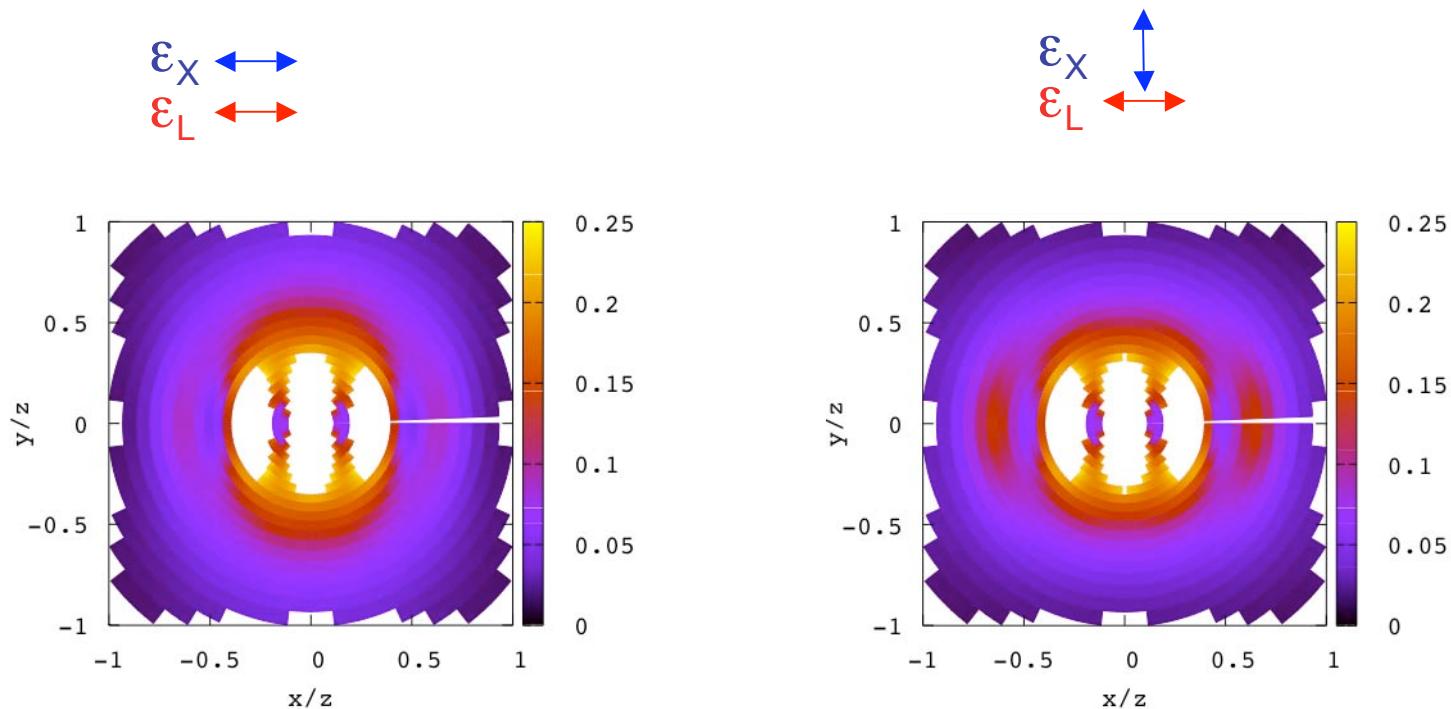


Calcs by Butch and Santra

Elastic scattering from Br_2



Polarization enhancement in diffractive imaging



Br_2 1 K
95 ps laser
1 ps x rays

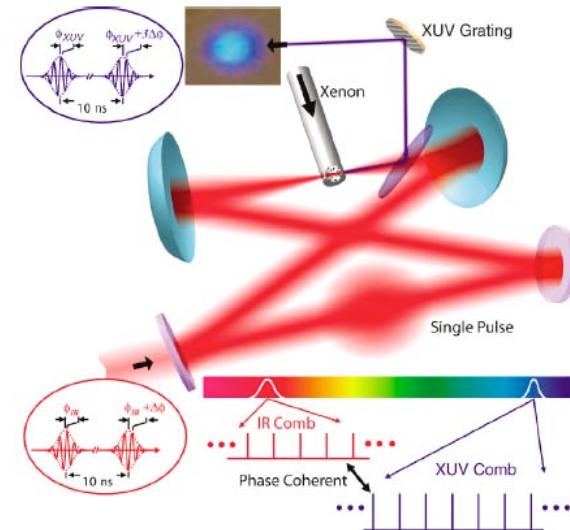
Future 2: Higher repetition rate 1 kHz to 6.5 MHz

Passive enhancement cavity
Jun Ye et al.

Coherent addition of successive pulses



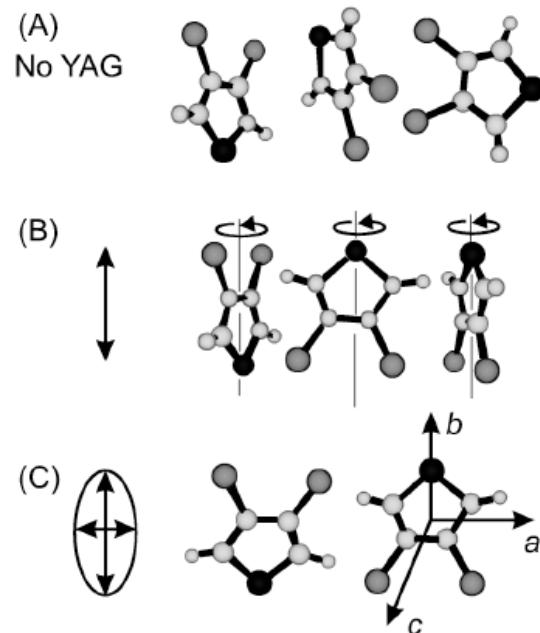
50 kHz - 10 MHz
10-100 ps, 200 μ J @ 1 μ m
10 Watts average power



Pulse energy 500x - >1 mJ
 $3 \times 10^{14} \text{ W/cm}^2$ Hartl et al., OL (2007)

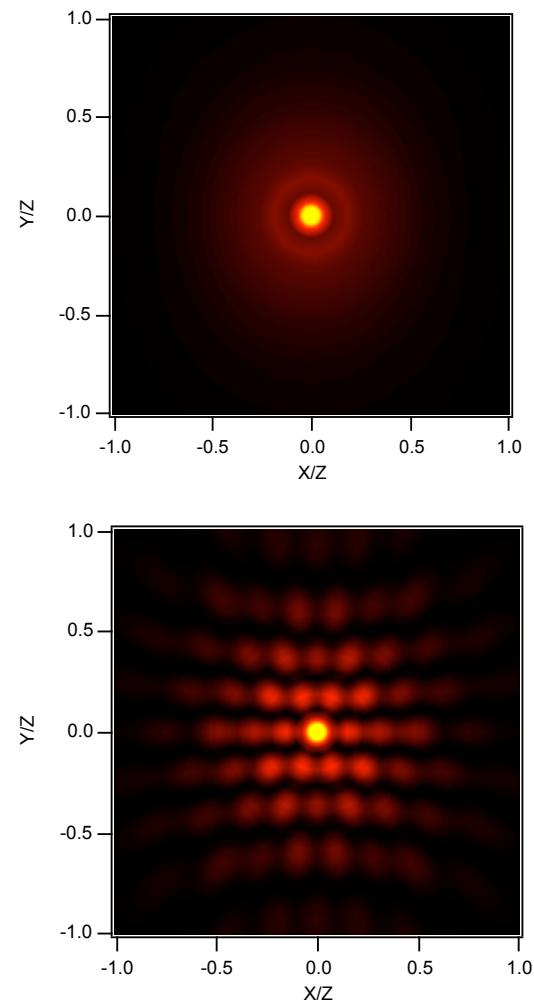
Future 3: 3D - alignment

3-D alignment w/elliptically polz'd fields
3,4 dibromothiophene



J.J. Larsen et al., PRL 85, 2470 (2000)

Field-free 3D Alignment - K. Lee et al. PRL (2006)

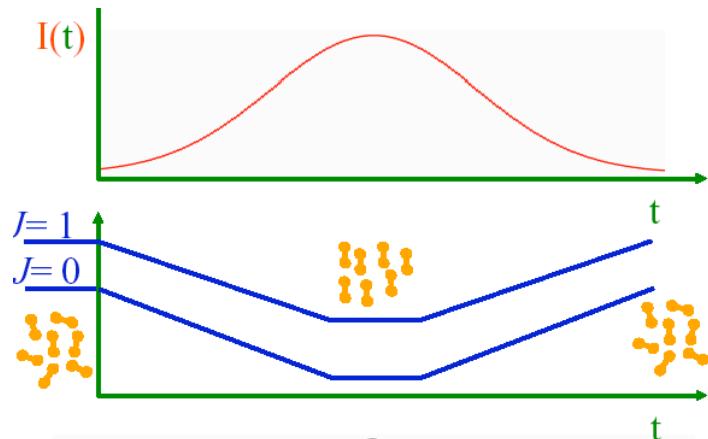


Adiabatic vs field-free alignment

- Assignment of symmetries of near edge resonances
- Behavior of molecules as fcn of strength of aligning potential
(Aligned molecules aid biomolecule structure determination)
- Photoelectron angular distributions from fixed-in-space molecules
- X-ray damage mechanisms
- Dynamics of laser-controlled molecular motion: fs & Angstrom

Adiabatic alignment

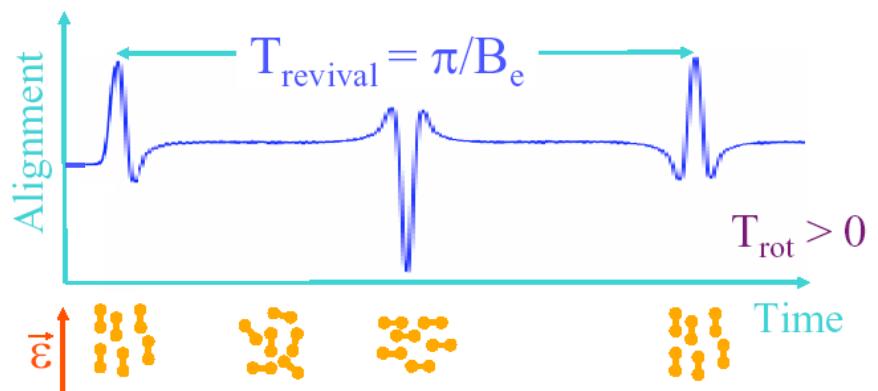
$$\tau_{\text{laser}} > \tau_{\text{rot}}$$



Friedrich & Herschbach, PRL 74, 4623 (95)

Field-free alignment

$$\tau_{\text{laser}} < \tau_{\text{rot}}$$



T. Seideman, PRL 83, 4971 (99)

Arbitrary pulse length laser aligning pulses Dynamical evolution of molecular rotational wavepacket

*Solve the time-dependent Schrodinger equation
Linear rotor/symmetric top + nonresonant linearly polarized laser field*

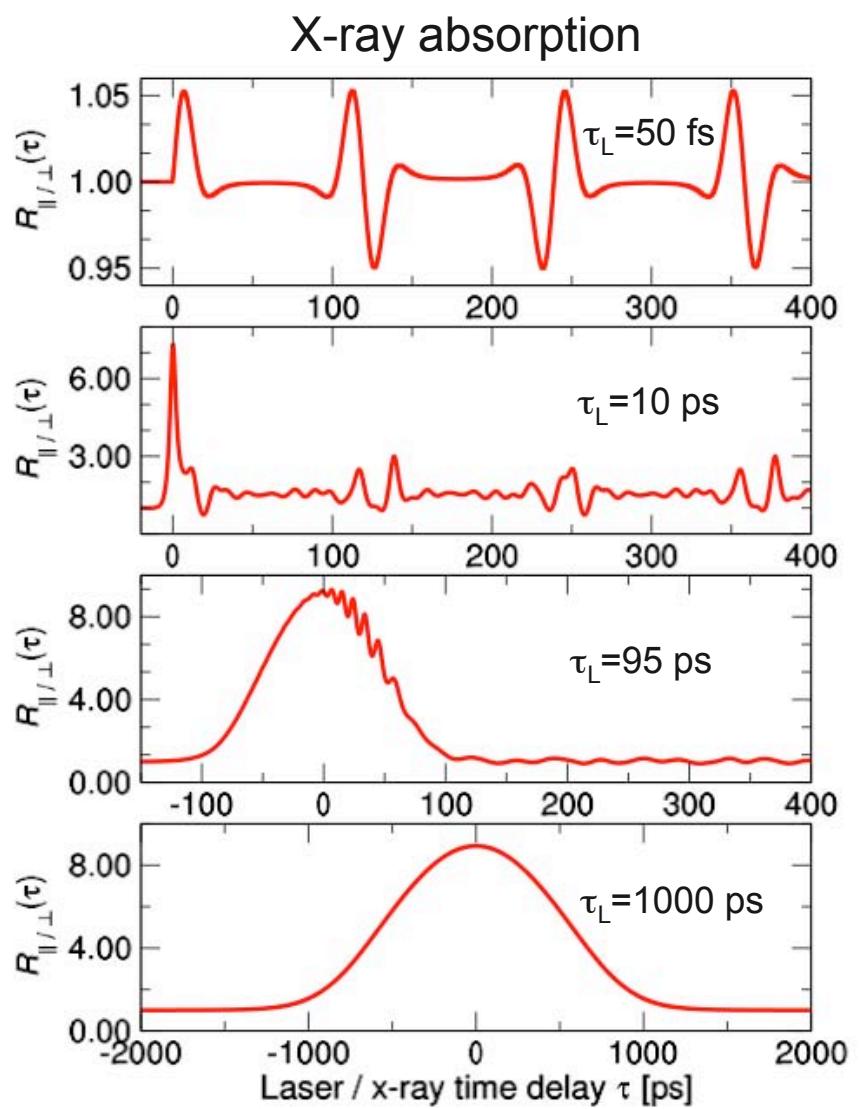
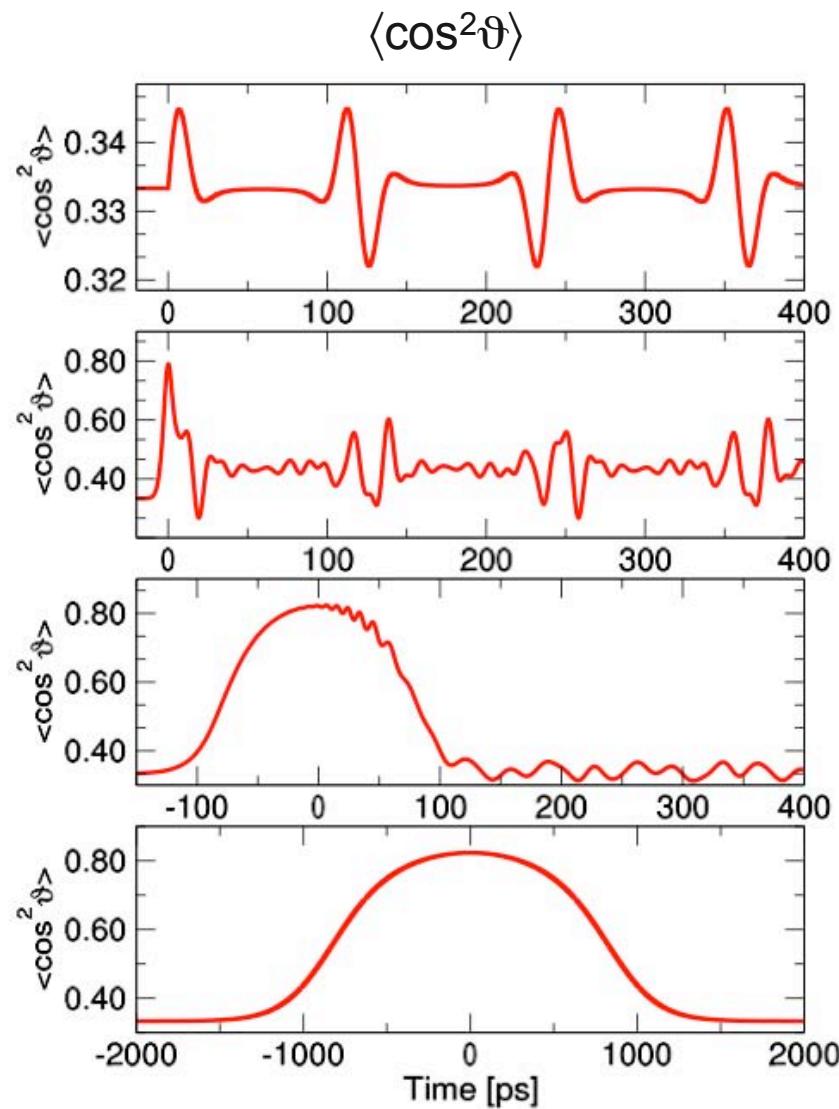
$$\vec{E}(t) = \hat{\epsilon} E_0 f(t) \cos \omega t,$$

$$V_{\text{int}} = -\frac{1}{2} E(t)^2 (\Delta \alpha \cos^2 \theta + \alpha_{\perp}),$$

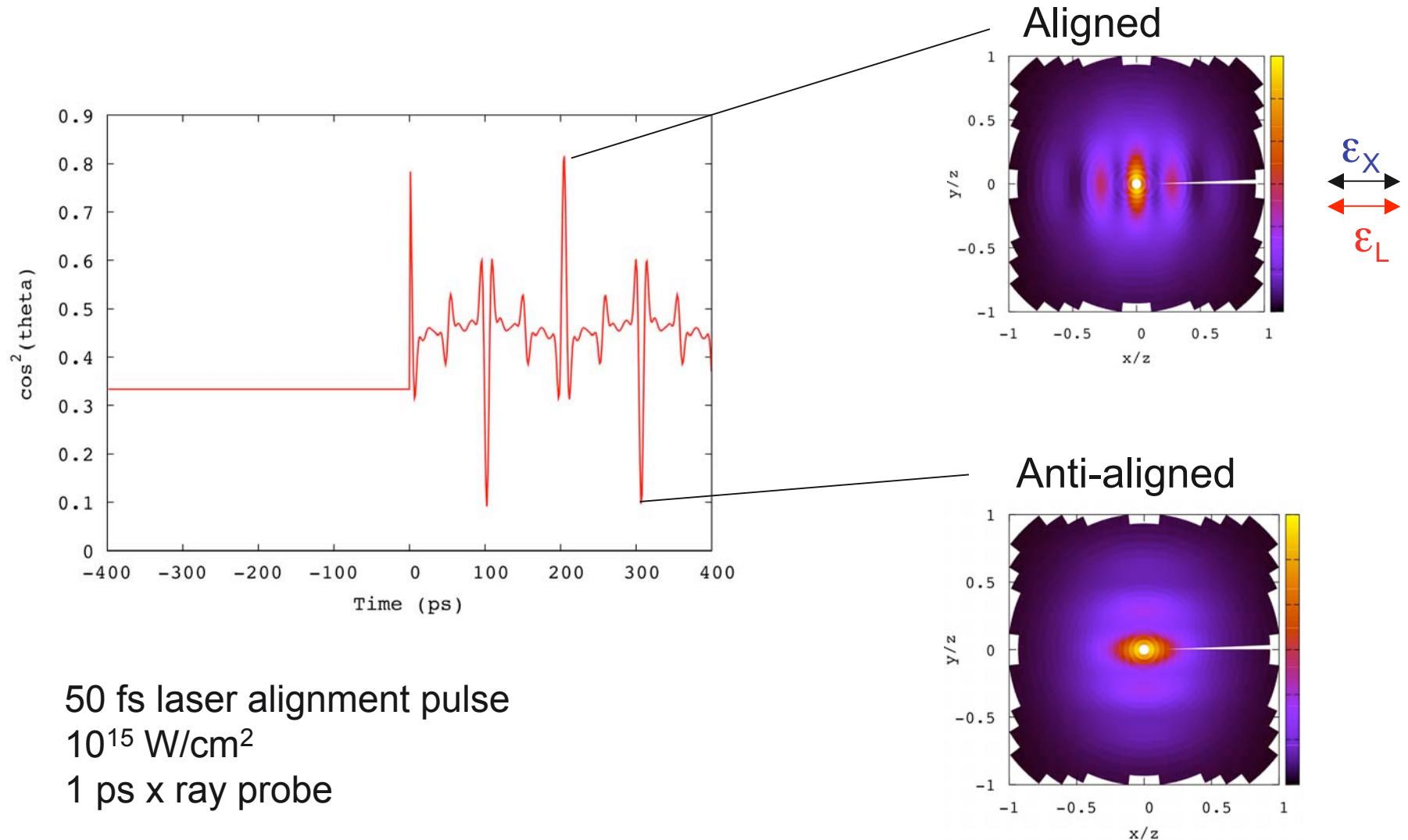
$$|\psi_{J_i M_i}\rangle = \sum_{J \geq |M_i|} F_{J_i J}(t) |JM_i\rangle,$$

Torres et al. PRA (2005)

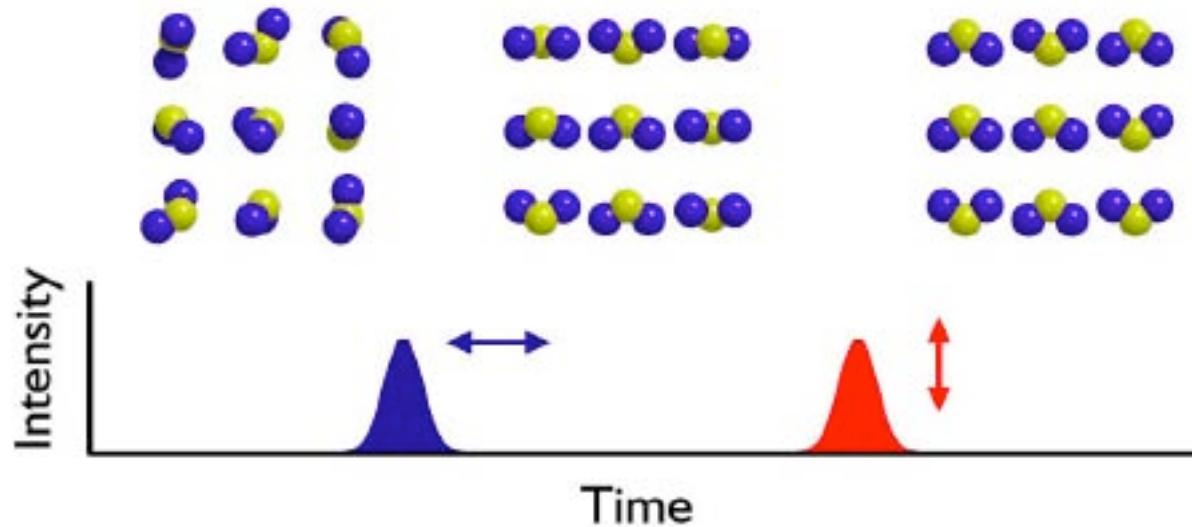
Laser alignment - from adiabatic to field-free



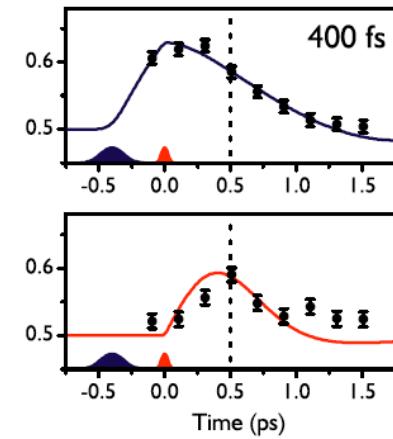
Coherent diffraction from field-free aligned Br_2 molecules



Future 4: Field-free 3D alignment



SO_2 : $\tau_1 = 180 \text{ fs}$, $\tau_2 = 50 \text{ fs}$, $\Delta\tau = 400 \text{ fs}$
Alignment duration $\sim 1 \text{ ps}$



Why we want 1-ps x-rays - a few AMO applications

- *Coherent diffraction imaging of molecules at higher field strength monitor laser-induced distortions with sub-Ångstrom resolution current 10^{12} W/cm² @ 100 ps to 10^{14} W/cm² @ 1 ps test ab initio understanding of dynamic polarizabilities non-perturbative multiphoton effects*
- *Coherent diffraction imaging of molecules in a field-free environment*
- *Dynamics of laser-controlled motions: rotational & vibrational (phonon)*
- *Photoionization dynamics from aligned molecules*